

Ultra-short, off-resonant, strong excitation of two-level systems

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We present a model describing the use of ultra-short strong pulses to populate the excited level of a two-level quantum system. In particular, we study an off-resonance excitation with a few cycles pulse which presents a *smooth phase jump* i.e. a change of the pulse's phase which is not step-like, but happens over a finite time interval. A numerical solution is given for the time-dependent probability amplitude of the excited level. The enhancement of the excited level's population is optimized with respect to the shape of the phase transient, and to other parameters of the excitation pulse.

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I. INTRODUCTION

Ultra-strong pulses with intensities of the order of 10^{15} W/cm², and duration of the order of attoseconds, with just few optical cycles, are feasible with present day technology (see e.g. [1–5]). This technological development has been motivated by the large number of possible applications, several of which rely on coherent population transfer techniques. A partial list of such applications is: stimulated Raman adiabatic passage (STIRAP) [6–9], adiabatic rapid passage (ARP) [10], Raman chirped adiabatic passage (RCAP) [11, 12], temporal coherent control (TCC) [13, 14], coherent population trapping [15, 16], optical control of chemical reactions [17, 18], electromagnetically induced transparency (EIT) [15, 19–22], efficient generation of XUV radiation [23–26], coherent Raman umklappscattering [27], breakdown of dipole blockade obtained driving atoms by phase-jump pulses [28]. Moreover, recently two schemes for efficient and fast coherent population transfer have been presented [29], which use chirped and non-chirped few-cycles laser pulses. Another recent application [30] presents high-order harmonic generation obtained with laser pulses with a π -phase jump. Finally, the field of quantum information processing benefits from these results, since many qubit realizations rely on precise quantum levels manipulation [31–34].

The presence of few optical cycles in the pulse gives a constant phase difference between the carrier wave and the pulse shaped envelope [35], in contrast with many cycle pulses [28, 36, 37]. Moreover, optimizing the pulses parameters is proven to enhance the excited state population [38] or optimizing coherence in two-level systems (TLSs) [39]. In previous works we have already presented an analytical solution for the dynamics of a TLS excited with pulses of arbitrary shape and polarization [40, 41]. Since in the model we present the change rate of levels' populations within a single optical cycle

is not negligible, the rotating-wave approximation can't be used. In other words in the present model we can't neglect the contribution of the counter-rotating terms in the Hamiltonian [40, 41].

In a previous work [42] we have presented a similar model, representing the interaction of a TLS with few-cycle pulses, where at time $t = t_0$ the phase of the carrier wave jumps of an amount ϕ , this jump being sharp and step-like. In that work, numerical analysis of the analytic model has lead to an enhancement by a factor of $10^6 - 10^8$ in the population transfer, with the optimal phase jump of $\phi = \pi$ and the optimal time coincident with the peak of the envelope. In the present work we improve that model, considering a *smooth phase change*, i.e. not step-like but happening over a finite interval of time. This new model more closely describes a realistic experimental scenario.

The pulse is characterized by: Rabi frequency Ω_0 , pulse width τ , carrier frequency ν , phase jump amplitude ϕ , phase jump time t_0 and phase jump duration Δt . Moreover, we consider two *qualitative parameters*: the phase jump shape, and the pulse envelope shape.

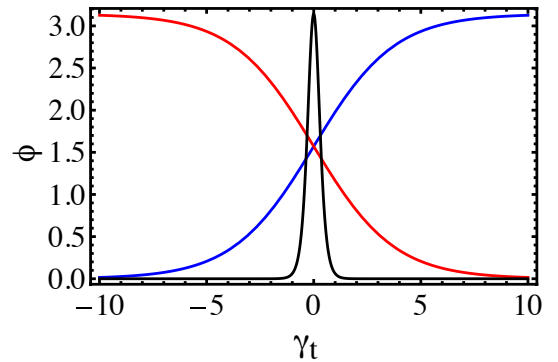


FIG. 1: The three functional shapes of the smooth phase jumps used: the red is a dropping hyperbolic tangent: $\phi(t) = (\pi/2)[1 - \tanh(5\alpha t)]$, the blue is a rising hyperbolic tangent: $\phi(t) = (\pi/2)[1 + \tanh(5\alpha t)]$, and the black one is a hyperbolic secant: $\phi(t) = (\pi/2)\text{sech}(\alpha t)$. In all the simulations the numerical normalized value is $\alpha = 0.265$

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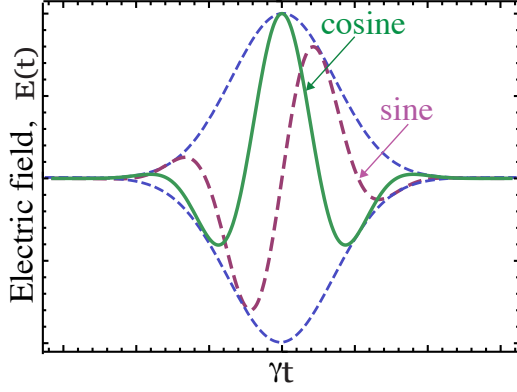


FIG. 2: Example of an excitation field with few oscillations and two different phases, thick solid(cosine) and dashed lines(sine), encompassed by a pulsed envelope (thick dashed line). Here the field is written in the form $E(t) = \mathcal{E}(t) \cos(\nu t + \phi_0)$ where $\phi_0 = 0$ for cosine and $\phi_0 = -\pi/2$ for sine pulse.

We present an analytical solution for the time evolution of the excited state's population, together with a numerical simulation. In the numerical simulation we use 3 functional shapes for the smooth phase jump: rising hyperbolic tangent, dropping hyperbolic tangent, and gaussian peak, (see figure 4), whereas for the envelope a gaussian peak has been used. Numerically optimizing the pulses parameters we have obtained enhancements for the population transfer of the order of 10^4 .

A. NUMERICAL SIMULATION AND DISCUSSION

Be $|a\rangle$ and $|b\rangle$ the states of a two-level atom (TLA), with energy difference $\hbar\omega$, and atomic dipole moment \wp . If we let this system interact with a classic field $E(t) = \mathcal{E}(t)\cos\nu t$, the equations of motion for the relative wavefunctions are [43]:

$$\dot{C}_a = i \frac{\wp \mathcal{E}(t)}{\hbar} \cos(\nu t) e^{i\omega t} C_b, \quad (1a)$$

$$\dot{C}_b = i \frac{\wp^* \mathcal{E}(t)}{\hbar} \cos(\nu t) e^{-i\omega t} C_a, \quad (1b)$$

where $\Delta = \omega - \nu$ is the detuning from resonance. Similarly to [42], defining $f(t) = C_a(t)/C_b(t)$ and $\Omega(t) = \wp \mathcal{E}(t)/\hbar$, we have the following Riccati equation:

$$\dot{f} + i\Omega^*(t)\cos(\nu t)e^{-i\omega t}f^2 - i\Omega(t)\cos(\nu t)e^{i\omega t} = 0. \quad (2)$$

The approximate solution for Eq. (2), in terms of the tip angle θ is given as in [40]

$$f(t) = i \int_{-\infty}^t dt' \left\{ \left[\frac{d\theta(t')}{dt'} - \theta^2(t') \frac{d\theta^*(t')}{dt'} \right] \times \exp \left[2 \int_{t'}^t \theta(t'') \dot{\theta}^*(t'') dt'' \right] \right\}, \quad (3)$$

where the tip angle $\theta(t)$ has been defined as

$$\theta(t) = \int_{-\infty}^t \Omega(t') \cos(\nu t') e^{i\omega t'} dt' \quad (4)$$

from which we have $|C_a(t)| = |f(t)| / \sqrt{1 + |f(t)|^2}$. What is of interest is the asymptotic behavior of $|C_a(\infty)|$. In [42] is shown good agreement between the analytical and a numerical simulation. To introduce the phase jump, we can write the Rabi frequency as

$$\Omega(t) = \Omega_0(t) \cos \nu t e^{i\omega t} e^{i\phi(t)} \quad (5)$$

and then, using the same method as in [42], we can obtain an approximated analytic solution for the Riccati equation (2):

$$f(t) = i \int_{-\infty}^t dt' \Phi(t') \exp \left[2 \int_{t'}^t \zeta(t'') dt'' \right], \quad (6)$$

The approximate analytical solution is in good agreement with the numerical simulation obtained by directly solving the coupled differential Eq.(1). From Fig.(3) we see that even for complex phase function the agreement is good. For the sake of completeness, we have added an appendix in which we show the strength of this approach beyond standard TLS. Indeed the Riccati equation approach gives a closed compact form from which

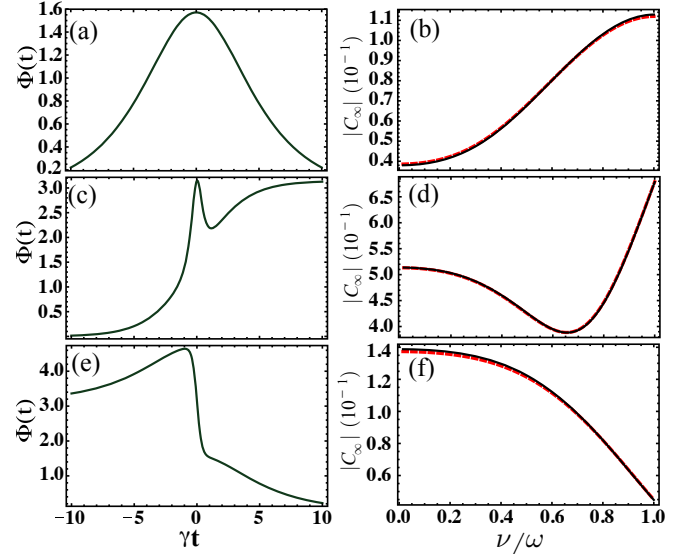


FIG. 3: Population left on the upper-level $|a\rangle$ (b,d,f) as a function of ratio of the carrier-frequency(ν) of the excitation pulse and atomic transition frequency(ω_c), in the long time limit $t \gg \tau$, for corresponding phase jump function (a,c,e). Here the dashed line if the numerical simulation of Eq.(1) and the solid line is the approximate solution given by Eq.(6). For the excitation pulse we have used the form $\Omega_0(t) = A e^{-a^2 t^2} e^{i\phi(t)}$, and the phase functions have the following forms: (a) $\phi(t) = (\pi/2) \text{sech}[at]$, (c) $\phi(t) = \pi/2(\text{sech}[10at] + (1 + \tanh[at]))$, (e) $\phi(t) = \pi/2(\text{sech}[at] + (1 - \tanh[10at]))$. For numerical simulations we chose $A = 0.035\omega$, $\alpha = 0.265\gamma$ and $\gamma = 1.25\omega$ where $\omega = (2\pi) 80 \text{ GHz}$

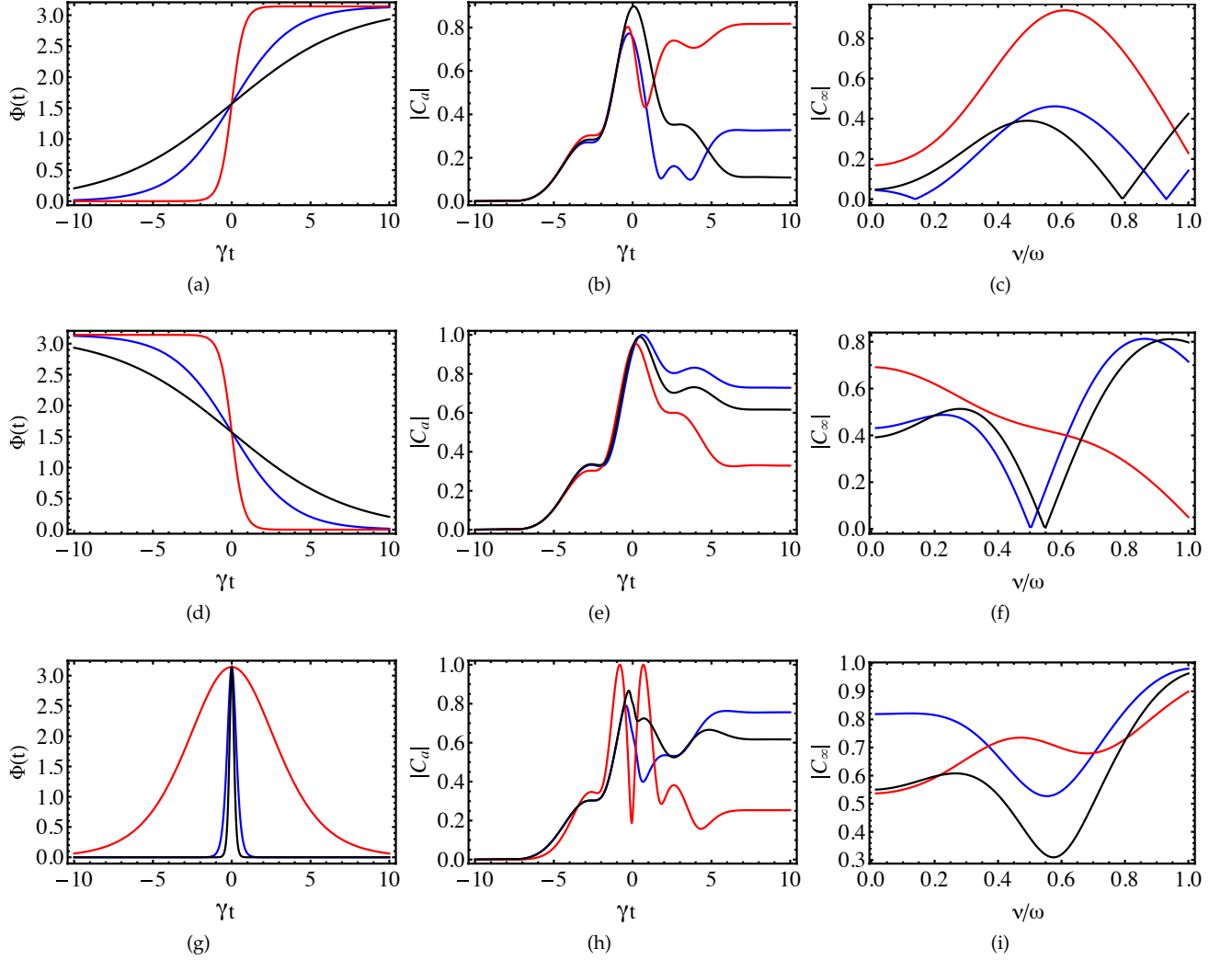


FIG. 4: In this figure we present the results of the numerical analysis. Each of the three rows of plots refers to a different functional shapes of the smooth phase jump (phase change function). For each row we have a plot of the smooth phase jump, a plot of the excited state's population as function of time, and a plot of the excited state's population left after the pulse is gone as a function of the normalized excitation frequency. Similarly to figure 3, the functional shape of the excitation pulse is $\Omega_0(t) = Ae^{-a^2 t^2} e^{i\phi(t)}$. Moreover, for the plots of the excited state's population as function of time we have used the numerical value of $\nu/\omega = 0.75$. Phase change of the form (a) $\phi(t) = (\pi/2)[1 + \tanh(\alpha_1 t)]$, with three different values of α_1 : red (steeper) $\alpha_1 = 5\alpha$; blue (in-between) $\alpha_1 = \alpha$; black (smoother) $\alpha_1 = 0.5\alpha$. (b) Corresponding behavior of the excited level population $|C_d(t)|$. (c) Asymptotic value of the excited state population, as a function of the “resonance ratio” (excitation's frequency divided by transition's frequency) for this form of the phase change. (d) Phase change of the form $\phi(t) = (\pi/2)[1 - \tanh(\alpha_1 t)]$, with (as in (a)) three different values of α_1 : red (steeper) $\alpha_1 = 5\alpha$; blue (in-between) $\alpha_1 = \alpha$; black (smoother) $\alpha_1 = 0.5\alpha$. (e) Corresponding behaviour of the excited level population $|C_d(t)|$. (f) Asymptotic excited population for this form of the phase change. (g) Phase change of the form: $\phi(t) = (\pi/2)\text{sech}^2(\alpha_1 t)$, α_1 : red (larger) $\alpha_1 = \alpha$; blue (in-between) $\alpha_1 = 10\alpha$; black (narrower) $\alpha_1 = 20\alpha$. (h) Corresponding behaviour of the excited level population $|C_d(t)|$. (i) Asymptotic excited population for this form of the phase change. The value used for α is $\alpha = 0.265$. For numerical simulations we chose $A = 0.04375\omega$, $\alpha = 0.265\gamma$ and $\gamma = 1.25\omega$ where $\omega = (2\pi) 80$ GHz

both the temporal and steady-state behavior of the two and three-level system can be obtained.

An interesting observation is that it is possible to rewrite the Rabi frequency in (5) as

$$\Omega(t) = \Omega_0(t) \cos \nu t e^{i[\omega t + \phi(t)]} \quad (7)$$

and then define $\tilde{\omega}(t) = \omega + \phi(t)/t$ and interpret this as a

modulation of the atomic frequency, instead of a modulation of the excitation. Experimentally this can be realized in several ways, e.g. using modulated Zeeman or Stark effect.

Now we move to discuss our numerical simulation of the dynamics of the two-level atom interacting with ultra-short, off-resonant and gradually changing phase

$\phi(t)$. We have performed numerical solution of the Riccati equation, using different types of phase change (smooth phase jump) functions. The result of this numerical analysis is shown in figure 4. The goal of this study is to find the best phase change which allows for the best coupling (most efficient energy exchange) of the excitation pulse with the excited state.

In figure 4 we present the results of the numerical analysis. Each of the three rows of plots refers to a different functional shapes of the smooth phase jump (phase change function). For each row we have a plot of the smooth phase jump, a plot of the excited state's population as function of time, and a plot of the excited state's population left after the application of the pulse as a function of the normalized excitation frequency. Similarly to figure 3, the functional form of the excitation pulse is $\Omega_0(t) = Ae^{-\alpha^2 t^2} e^{i\phi(t)}$. Moreover, for the plots of the excited state's population as function of time we have used the numerical value of $\nu/\omega = 0.75$.

For this numerical simulation we have considered the following three phase functions (a)(b)(c): $\phi(t) = (\pi/2)[1 + \tanh(\alpha t)]$, (d)(e)(f): $\phi(t) = (\pi/2)[1 - \tanh(\alpha t)]$ and (g)(h)(i) $\phi(t) = (\pi/2) \text{sech}^2(\alpha t)$. We can see how the phase change duration Δt , i.e. the steepness of the $\phi(t)$ function, has not an unique effect on the excited population, and depends on the general shape of the phase change. In particular, it is worth noting that for ascending and descending phase changes built on the $\tanh(t)$ function the effect of the steepness is opposite. We can observe a global behaviour which relates the characterizing parameters of the phase change with the amplitude of the population of the excited state. Qualitatively, for the ascending hyperbolic tangent we observe that by increasing the slope the population increases. On the other hand, for the descendent hyperbolic tangent the effect of this parameter is reversed: decreasing the slope of phase change leads to a decrease of the population. We remark that these behaviors are only global, and are reversed for some small ranges of frequencies. As an example, in plot 4.(f), for low ranges of laser frequencies, by decreasing the slope we increase the population, which is opposite of the behavior observed for higher frequencies. For the peaked shape ($\text{sech}(t)$), no general behaviors are observed. However, for the intermediate range of frequencies it can be observed a link between the increasing of the pulse width and the increase of the population.

II. CONCLUSION

To conclude, here we report our analytical and numerical results to show the effect of smooth phase jump on the dynamics (both transient and steady-state). We observed that the temporal profile of the phase jump function $\phi(t)$ has a profound effect on the excited state population $|a\rangle$. The two-level considered here can be the Zeeman-sublevels and the ultra-short (few to multi-

cycle) pulse would be in the radio-frequency regime which has been reported in[44–46]. For an optimized phase function (within the set of parameters considered here), we were able to observe an enhancement of 10^4 in the population transfer. Such enhancement is seen in Fig. 4(f) (blue curve) which takes the value of $|C_\infty| = 0.002251$ at $\nu/\omega = 0.5$ and phase function is $\phi(t) = (\pi/2)[1 - \tanh(\alpha t)]$ where $\alpha = 0.265$. When $\alpha_1 = 5\alpha = 1.325$ the excited state population is enhanced by factor of $\sim 2 \times 10^2$. Similar enhancement is also observed for the phase function $\phi(t) = (\pi/2)[1 - \tanh(\alpha t)]$ at frequency $\nu/\omega \sim 0.9$ (see Fig. 4c). We not only can enhance excitation but for the same phase function and other choice of the parameter α , we can also suppress it. For example at near resonant excitation (see Fig. 4(f)), the excited state population can be suppressed by ~ 15 -fold when $\alpha = 1.325$ (red curve) in comparison to $\alpha_1 = 0.5\alpha$ (black curve). Such control over excited state dynamics using smooth phase jump as an external parameter can be useful in microwave controlled Raman[47, 48], EIT with superstructures[49] to name a few while on the other hand with recent proposal on coherence-enhanced spaser[50] and propagating surface plasmon polaritons[51] also viable areas to explore phase effects. The approximate analytical solution are in excellent agreement for both delta function[42] or smooth phase jump considered here. We also extended this approach beyond two-level atom to three-level in lambda configuration.

III. ACKNOWLEDGEMENT

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Appendix A: Analytical solution for three-level atom

Motivation to add an appendix on the approximate analytical solution for three-level system in lambda configuration is to enlighten the strength of the method used to find the solution for two level atom with and without phase jumps. For the sake of simplicity we will consider constant phase ϕ . Let us consider a three-level atom(ThLA) in Λ configuration [see Fig. 5 inset]. The transition $a \leftrightarrow c$ is driven by the field Ω_2 , while the field Ω_1 couples the $a \leftrightarrow b$ transition. For the time scale considered in this problem, we have neglected any decays (radiative and non-radiative). The equation of motion for the probability amplitudes for the states $|a\rangle$, $|b\rangle$ and $|c\rangle$ of the ThLA can be written as

$$\dot{C}_a(t) = i\tilde{\Omega}_1(t)C_b(t) + i\tilde{\Omega}_2(t)C_c(t) \quad (\text{A1})$$

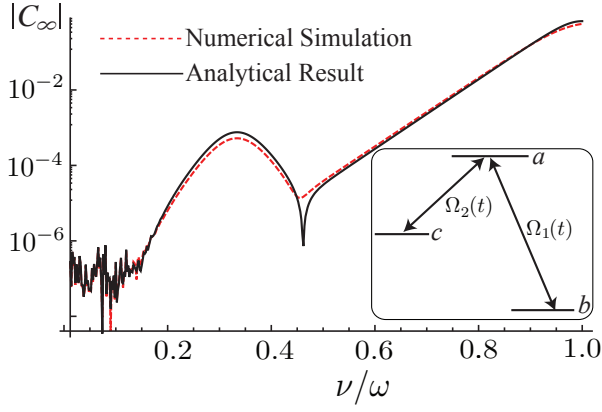


FIG. 5: Numerical (red dotted line) and analytical (blue solid line) solutions of the amplitude of the state $|a\rangle$ after long time in function of ν/ω for the laser pulse envelopes $\Omega_1(t) = \Omega_2(t) = \Omega_0 \text{sech}(\alpha t)$. For numerical simulation we chose $\Omega_0 = .04\omega$, $\alpha = 0.075\omega$, $\omega_{ab} = \omega_{ac} = \omega = 1$.

$$\dot{C}_b(t) = i\tilde{\Omega}_1^*(t)C_a(t) \quad (\text{A2})$$

$$\dot{C}_c(t) = i\tilde{\Omega}_2^*(t)C_a(t) \quad (\text{A3})$$

where $\tilde{\Omega}_j(t)$ is defined as the effective Rabi frequencies

$$\tilde{\Omega}_j(t) = \Omega_j(t) \cos(\nu_j t) e^{i\omega_j t}; \quad j = 1, 2 \quad (\text{A4})$$

To solve for $C_a(t)$ and $C_c(t)$ let us define

$$f(t) = \frac{C_a(t)}{C_b(t)}, \quad g(t) = \frac{C_c(t)}{C_b(t)} \quad (\text{A5})$$

In terms of $f(t)$ and $g(t)$ Eqs.(A1, A2, A3) reduces to

$$\dot{f}(t) + i\tilde{\Omega}_1^* f^2(t) = \tilde{\Omega}_1 + i\tilde{\Omega}_2 g(t) \quad (\text{A6})$$

$$\dot{g}(t) + i\tilde{\Omega}_1^* f(t)g(t) = i\tilde{\Omega}_2^* f(t) \quad (\text{A7})$$

In order to solve these equations we extended the method developed[40, 41]. By neglecting the non-linear term $f^2(t)$ and the term $\propto g(t)$ in Eq.(A6) we can solve for $f_1(t)$ as

$$f_1(t) = i \int_{-\infty}^t \tilde{\Omega}_1 dt' \quad (\text{A8})$$

Similarly by neglecting the term $\propto g(t)$ in Eq.(A7) we can solve for $g_1(t)$ as

$$g_1(t) = - \int_{-\infty}^t \tilde{\Omega}_2^*(t') \theta_1(t') dt' \quad (\text{A9})$$

where the tip angle $\theta_1(t)$ is defined as

$$\theta_1(t') = \int_{-\infty}^{t'} \tilde{\Omega}_1(t') dt' \quad (\text{A10})$$

Next let us write the non-linear term in Eq.(A6) as

$$f^2(t) = [f(t) - f_1(t)]^2 + 2f(t)f_1(t) - f_1^2(t) \quad (\text{A11})$$

Then Eq.(A6) can be written as

$$\dot{f}(t) + i\tilde{\Omega}_1^*(t) \{ [f(t) - f_1(t)]^2 + 2f(t)f_1(t) - f_1^2(t) \} = i\tilde{\Omega}_1(t) + i\tilde{\Omega}_2(t)g(t) \quad (\text{A12})$$

Let us assume that $g(t) \approx g_1(t)$ and we neglect $[f(t) - f_1(t)]^2$ [40] in this case we can write Eq.(A12) in term of the tip angles $\theta_1(t)$ and $\theta_2(t)$

$$\dot{f}(t) + i\dot{\theta}_1^*(t) \{ 2f(t)f_1(t) - f_1^2(t) \} = i\dot{\theta}_1(t) + i\dot{\theta}_2(t)g_1(t) \quad (\text{A13})$$

where

$$\theta_2(t') = \int_{-\infty}^{t'} \tilde{\Omega}_2(t') dt', \quad (\text{A14})$$

The analytical solution of the equation Eq.(A13) is then:

$$f(t) = e^{-a(t)} \int_{t_0}^t b(t') e^{a(t')} dt' \quad (\text{A15})$$

where

$$a(x) = 2i\dot{\theta}_1(t)f_1(t) \quad (\text{A16})$$

and

$$b(x) = i\dot{\theta}_1(t) + i\dot{\theta}_2(t)g_1(t) + i\dot{\theta}_1^*(t)f_1^2(t) \quad (\text{A17})$$

For $g(t)$ the solution can be obtain from Eq.(A7) where we use $f(t) \approx f_1(t)$:

$$\dot{g}(t) + i\dot{\theta}_1^*(t)f_1(t)g(t) = i\dot{\theta}_2^*(t)f_1(t) \quad (\text{A18})$$

which give us

$$g(t) = e^{-c(t)} \int_{t_0}^t D(t') e^{c(t')} dt' \quad (\text{A19})$$

where

$$c(x) = i\dot{\theta}_1^*(t)f_1(t) \quad (\text{A20})$$

and

$$D(x) = i\dot{\theta}_2^*(t)f_1(t) \quad (\text{A21})$$

In Fig. 5 we have plotted the Numerical (red dotted line) and analytical (blue solid line) solutions of the amplitude of the state $|a\rangle$ after long time in function of ν/ω_c for the laser pulse envelopes $\Omega_1(t) = \Omega_2(t) = \Omega_0 \text{sech}(\alpha t)$ with $\Omega_0 = .04\omega$, $\alpha = 0.075\omega$, $\omega_{ab} = \omega_{ac} = \omega = 1$. We see that the approximate analytical solution matches well with the numerics under the parameters considered here. Extension of this methodology to Schrodinger equation[52, 53] and position dependent mass Schrodinger (PDMSE) equation can be found in[54, 55].

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